Study of the influence of a spin-orbit exciton on the magnetic ordering in Sr_2IrO_4

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Abstract. This study investigates an orbital exciton propagation in quasi-two-dimensional Heisenberg antiferromagnet Sr_2IrO_4 by means of computer simulation. Ising and Heisenberg models are compared, taking in consideration magnetic interactions and exciton hopping. As the result of this simulation the spin structure factor for both models is calculated

1. Introduction

Sr_2IrO_4 is a $5d$ transition metal oxide. It is one of recently discovered Mott insulators and, while other $5d$ transition metal oxides with a similar structure are metallic, this compound does insulate. Sr_2IrO_4 shares several distinctive features with cuprates which are characteristic for superconductivity. It makes Sr_2IrO_4 an object of great interest as a possible high temperature superconductor [1, 2].

Sr_2IrO_4 can be represented as a quasi-two-dimensional Heisenberg antiferromagnet with isospin 1/2. Here isospin is the effective total angular momentum of occupied energy states of $5d$ configuration of Ir as a good quantum number [3, 4].

Unlike the Mott gap in 3d transition metal oxides caused by strong Coulomb repulsion, the Mott gap in Sr_2IrO_4 has a different nature. Under the O_h symmetry $5d$ states of Ir split into the t_{2g} triplet and e_g doublet. In the strong spin-orbit coupling limit the t_{2g} bang splits into a full quartet with isospin 3/2 and a hall-filled doublet with isospin 1/2 (figure 1). Even a relatively small Coulomb repulsion opens up the Mott gap near E_F and splits the $S = 1/2$-manifold into the upper (UHB) and lower Hubbard bands (LHB), shown in figure 2 [3].

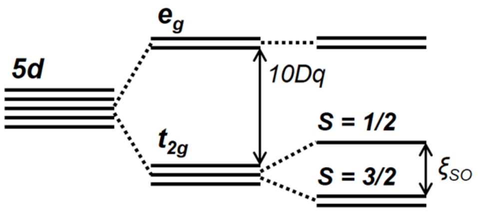

Figure 1. Splitting of $5d$ manifold by the crystal field and spin-orbit coupling [3].

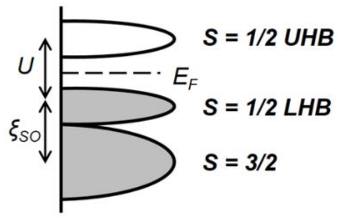

Figure 2. Schematic energy diagram for a $5d$ configuration with the strong spin-orbit coupling and Coulomb repulsion [3].

In experimental spectra of Sr_2IrO_4 one can observe modes of a charge-neutral quasi-particle. This quasi-particle is referred to as 'spin-orbit exciton'. It propagates through the lattice misaligning spins and changing the magnetic ordering [1, 2]. Such an excitation can be induced by X-rays [1] or Rhdoping [5]. Some researchers expect that the spin-orbit exciton propagation in Sr_2IrO_4 can lead to high temperature superconductivity.

In this work we use computer simulation to investigate the propagation of a single spin-orbit exciton through Sr_2IrO_4. The magnon dispersion in this compound is well described by an antiferromagnetic Heisenberg model with isospin one-half moments on a square lattice [2]. For modelling we use a Heisenberg model and a simplier Ising model.

We have developed a program to model a single spin-orbit exciton motion through antiferromagnetically ordered lattice to qualitatively evaluate changes in the system's characteristics. Upon the obtained results we argue that a single spin-orbit exciton propagation through Sr_2IrO_4can be modelled with the Ising model instead of the Heisenberg one.

A rather big value of the spin-orbit coupling constant (ξ_{SO}~$0{,}4 - 0{,}5$ eV [2, 3]) allows to consider ground states in Ir ions as effectively reduced to a half-filled singlet system with isospin 1/2 (figure 3a). For an excited Ir ion we can draw such a scheme with one electron moved from the quartet to the doublet across the spin-orbit split. As now the doublet is filled, we can reduce the system to a quartet with three electrons (figure 3b). Its effective isospin is 3/2 [2].

a) b)

Figure 3. Schematic electronic configurations of a) an Ir ion in a ground state ($S = 1/2$) and b) in an excited state ($S = 3/2$).

The Hamiltonian of this modelling contains longitudinal and transverse exchange coupling and exciton hopping. We take in consideration only nearest neighbours. Any long-range interactions give much smaller value of exchange coupling constants and can be omitted.

The Hamiltonian with exciton hopping and anisotropic exchange coupling can be written as

$$\hat{H} = J_\parallel \sum_{\langle ij\rangle} S_i^z S_j^z + J_\perp \sum_{\langle ij\rangle} S_i^+ S_j^- - W \sum_{\langle ij\rangle} X_i^\dagger X_j, \quad (1)$$

where $J_\parallel$, $J_\perp$ are exchange coupling constants, S_i^z is an isospin projection operator, S_i^+, $S_j^-$are raising and lowering operators for isospin projections, $X_i^\dagger$, X_j denote exciton creation and annihilation operators. The hopping parameter $W = 2t^2/U$ is defined by the intraorbital Coulomb repulsion U and hopping integral t. $J_\perp = 0$ corresponds to the Ising model, while for the Heisenberg model $J_\perp = J_\parallel$. The Ising and Heisenberg models are special cases of the anisotropic exchange model.

J, the longitudinal and transverse exchange coupling constant, is the reduced unit of measurement in this modelling. J can be estimated as 60 meV [2], about 650 K. It is omitted in further notation.

2. Method

In order to model such a system we use Stochastic Series Expansion (SSE) [6] – one of quantum Monte-Carlo methods. Its outline is presented here.

The thermal expectation of an operator $\hat{A}$

$$\langle \hat{A} \rangle = \frac{1}{Z} Sp\{A e^{-\beta \hat{H}}\}, \quad (2)$$

where $Z = Sp\{e^{-\beta \hat{H}}\}$ is the partition function and β is the inverse temperature, is calculated as the expectation value in a combined space of operator sequences and spins. 'Subspace of spins' means that we flip spins in the system. Working in the subspace of operators means changing operator sequences, which are applied to the spin system.

The Hamiltonian could be represented as a sum of M non commutative terms

$$H = \sum_{i=1}^{M} \hat{H}_i, \quad [\hat{H}_i, \hat{H}_j] \neq 0 \tag{3}$$

In this approach we say that β is a small parameter, that is why this method works only for high temperatures. As H-terms do not commutate, the Taylor expansion can be written as a double sum with the product of operators in the sequence

$$e^{-\beta\hat{H}} = 1 - \beta\hat{H} + \frac{1}{2}(\beta\hat{H})^2 - \frac{1}{6}(\beta\hat{H})^3 + \cdots = \sum_{n=0}^{L} \sum_{\{C_n\}} \frac{(-\beta)^n}{n!} \prod_{j=1}^{n} \hat{H}_{l_j}, \tag{4}$$

where $\{C_n\}$is the subspace of operators and $l_j = 1, 2, \ldots, M$. Here the expansion is cut up to the L^{th} term

If we omit zero operators in the sequence, the formula for the expectation value of the operator $\hat{A}$ can be written as

$$\langle\hat{A}\rangle = \frac{1}{Z} \sum_{\alpha} \sum_{\{C_L\}} \frac{(-\beta)^{n(C_L)}[L-n(C_L)]!}{L!} \left\langle \alpha \middle| \hat{A} \prod_{j=1}^{L} \hat{H}_{l_i} \middle| \alpha \right\rangle, \tag{5}$$

where $n(C_L)$ denotes the number of nonzero operators in the operator sequence and $\{\alpha\}$ is the subspace of spin configurations.

The straightforward application of the SSE method to the considered system leads to generating a great number of non-physical states, so that many Monte-Carlo iterations are wasted. In order to avoid such configurations some modifications are required, which ensures that hopping operators of a certain sequence make the excitation move along a closed trajectory. According to selection rules we exclude generating those hopping operators which will surely turn the sequence weight into zero.

The algorithm we finally got calculates the spin structure factor

$$S(\boldsymbol{q}) = \frac{1}{N^2} \sum_{i,j} e^{i\boldsymbol{q}(\boldsymbol{r}_i - \boldsymbol{r}_j)} \langle \hat{S}_i^z \hat{S}_j^z \rangle \tag{6}$$

It shows what arrangements of spins are like. $S(\boldsymbol{q})$ is a surface in the reciprocal space. For antiferromagnetically ordered isospins it yields a peak at $\boldsymbol{q} = (\pi, \pi)$, any deviation from the antiferromagnetic ordering makes this peak lower and broader. When spins are ordered ferromagnetically it has a maximum at $\boldsymbol{q} = (0,\ 0)$.

3. Results

First, we have calculated spin structure factors for various temperatures to see what the spin structure factor is like and how the shape of its surface in the reciprocal space changes. For this comparison we consider a system with a localized exciton ($W = 0$) in figure 4 and a system with a propagating exciton (we set $W = 1$, i.e. the hopping parameter is equal to the exchange coupling constant). Here the values of spin structure factor along lines of symmetry in the reciprocal space are depicted.

At first sight it becomes obvious that $S(\boldsymbol{q})$ surfaces and their changes are very similar for both Heisenberg and Ising models. For some values of the system's parameters spin structure factor values for both models coincide within margin of error.

Both models demonstrate a finite peak at $\boldsymbol{q} = (\pi, \pi)$, which corresponds to the antiferromagnetic ordering. It broadens with temperature or hopping parameter. The shape of Heisenberg and Ising models' peaks are almost identical. The peak gets down slightly when the hopping parameter W changes from 0 to 1, but it still corresponds to the antiferromagnetic ordering, i.e. this amount of excitons per site with orbital excitation compared to exchange coupling is not enough to disorder antiferromagnetically ordered spins.

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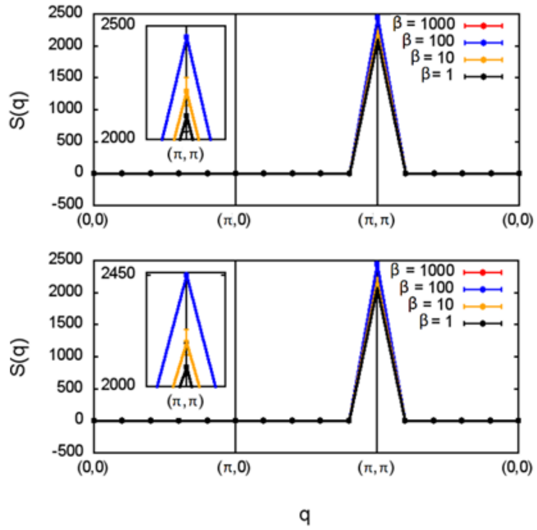

Figure 4. Spin structure factor $S(\boldsymbol{q})$ for the Heisenberg (upper) and Ising models (lower) at various temperatures for the 10x10 lattice at $W = 0$.

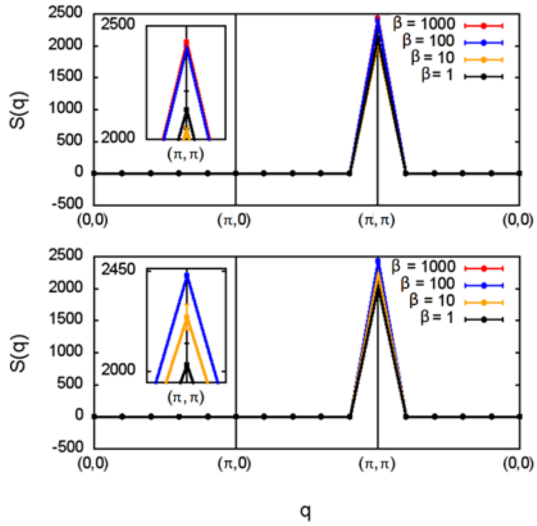

Figure 5. Spin structure factor $S(\boldsymbol{q})$ for the Heisenberg (upper) and Ising models (lower) at various temperatures for the 10x10 lattice at $W = 1$.

In these figures only results for the 10x10 lattice are presented. Other lattice sizes show similar behaviour in general.

Since we have some ideas of how the surface changes with temperature, we can keep an eye only on the point where the peak is expected to be – $\boldsymbol{q} = (\pi, \pi)$ – changes of the value at this point corresponds to the changes in the magnetic ordering. In figures 6 and 7 temperature dependencies for two lattice sizes are presented.

Temperature dependencies of spin factor peak for both models decline in a very similar way. The spin structure factor peak decreases with temperature in the systems for both lattice sizes. For the 4x4 lattice there's obviously a phase transition in the interval $10 < \beta < 40$. For the 10x10 lattice peak height undergoes a sharp decrease as well, but it does not look like the system has switched to the ferromagnetic or paramagnetic order with a peak height around 1000. For both lattice sizes bigger value of the hopping parameter corresponds to the lower temperature of the phase transition.

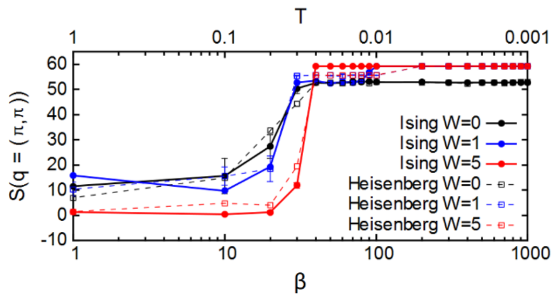

Figure 6. Temperature dependency of spin structure factor peak $S(\boldsymbol{q} = (\pi, \pi))$ for the 4x4 lattice.

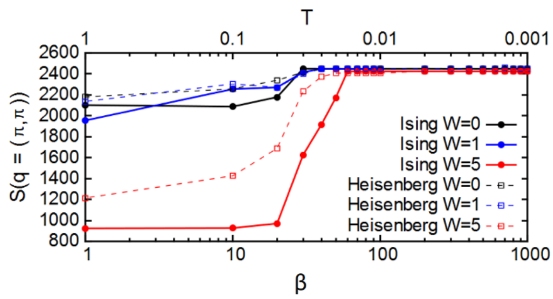

Figure 7. Temperature dependency of spin structure factor peak $S(\boldsymbol{q} = (\pi, \pi))$ for the 10x10 lattice.

Then we tracked how the system evolves when hopping is the varying parameter.

The peak of spin structure factor degrades with hopping for both temperatures in consideration (figures 8 and 9). At a low temperature the Ising and Heisenberg models give almost the same result, except for $W = 2$ (figure 8) – here for the Ising model we get the paramagnetic order while in the Heisenberg model the order is antiferromagnetic.

By the peak's shape and height at a higher temperature (figure 9) antiferromagnetic ordering can be expected at all values of the hopping parameter. In this comparison the Ising and Heisenberg models show themselves very similar – the spin structure factor heights go down with hopping in similar ways.

One can draw an analogy between the influence of spin-orbit exciton propagation (figures 8 and 9) and the influence of heat fluctuations (figures 4 and 5).

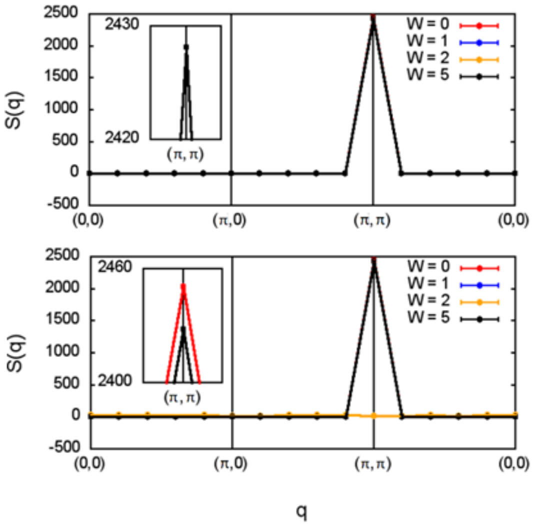

Figure 8. Spin structure factor $S(\boldsymbol{q})$ for the Heisenberg (upper) and Ising models (lower) at various hopping parameters W for the 10x10 lattice at $\beta = 1000$.

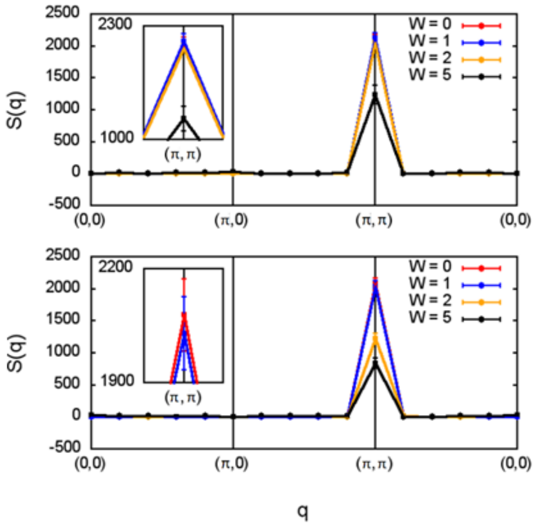

Figure 9. Spin structure factor $S(\boldsymbol{q})$ for the Heisenberg (upper) and Ising models (lower) at various hopping parameters W for the 10x10 lattice at $\beta = 100$.

In figures 10 and 11 peak heights decrease with hopping at high temperatures and remains almost constant at $\beta = 1000$. One can notice that Heisenberg and Ising models yield the same character of losing the antiferromagnetic order with hopping. But at $W = 0$ the Ising model gives peak heights close to 0, though earlier calculations had a high peak (figure 4). It shows that at these parameters ($W = 0$) the Ising model is unstable and can be driven to the paramagnetic ordering.

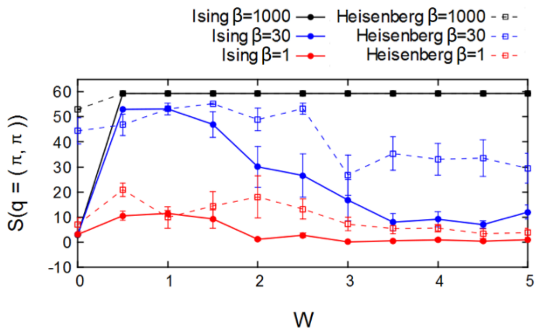

Figure 10. Hopping parameter dependency of spin structure factor peak $S(\boldsymbol{q} = (\pi, \pi))$ for the 4x4 lattice.

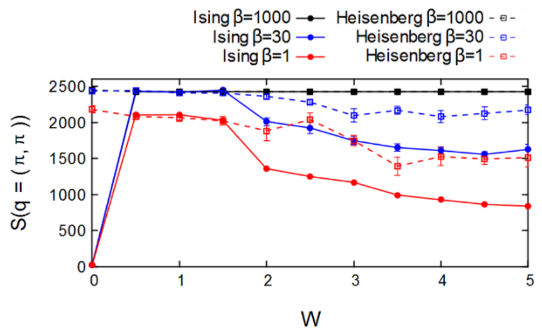

Figure 11. Hopping parameter dependency of spin structure factor peak $S(\boldsymbol{q} = (\pi, \pi))$ for the 10x10 lattice.

4. Conclusion

Both models demonstrate the exciton-induced system getting disordered with spin-orbit exciton hopping and heat fluctuations. Some of results for these two models agree within margin of error. But the Ising model proves to be unstable yielding extremely different results when calculations are repeated. Yet at other values of the system parameters the Ising model qualitatively is rather close to the Heisenberg model's results and experimental and theoretical data.

As it was expected, with exciton hopping an exchange frustration appears. Such frustrations can be observed in both Heisenberg and Ising models, the scales of these frustrations are very much alike. That makes the Heisenberg and Ising models similar for simulation of a spin-orbit exciton propagation.

Thus, for modelling a single spin-orbit exciton propagation in Sr_2IrO_4 the Heisenberg model can be substituted with the Ising model, though with some limitations.

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